Supplementary Information

Nano-FTIR spectroscopy reveals SiO_2 densification within fs-laser induced nanogratings

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Abstract

This study explores the structural transformations induced by femtosecond (fs) laser inscriptions in glass, with a focus on Type II modifications (so called nanogratings), crucial for advanced optical and photonic technologies. Our novel approach employs scattering-type scanning near-field optical microscopy (s-SNOM) and the synchrotron radiation nanoscale Fourier-transform infrared spectroscopy (nano-FTIR) to directly assess the nanoscale structural changes in the laser tracks, potentially offering a comprehensive understanding of the underlying densification mechanisms. The results reveal the first direct nanoscale evidence of densification driven by HP-HT within fs-laser inscribed tracks, characterized by a significant shift of the main infrared (IR) vibrational structural band of silica glass. It reveals moreover a complex interplay between Type I and Type II modifications.

Keywords: fs-laser writing, type II modifications, nanogratings, s-SNOM, Nano-FTIR spectroscopy, silica glass densification

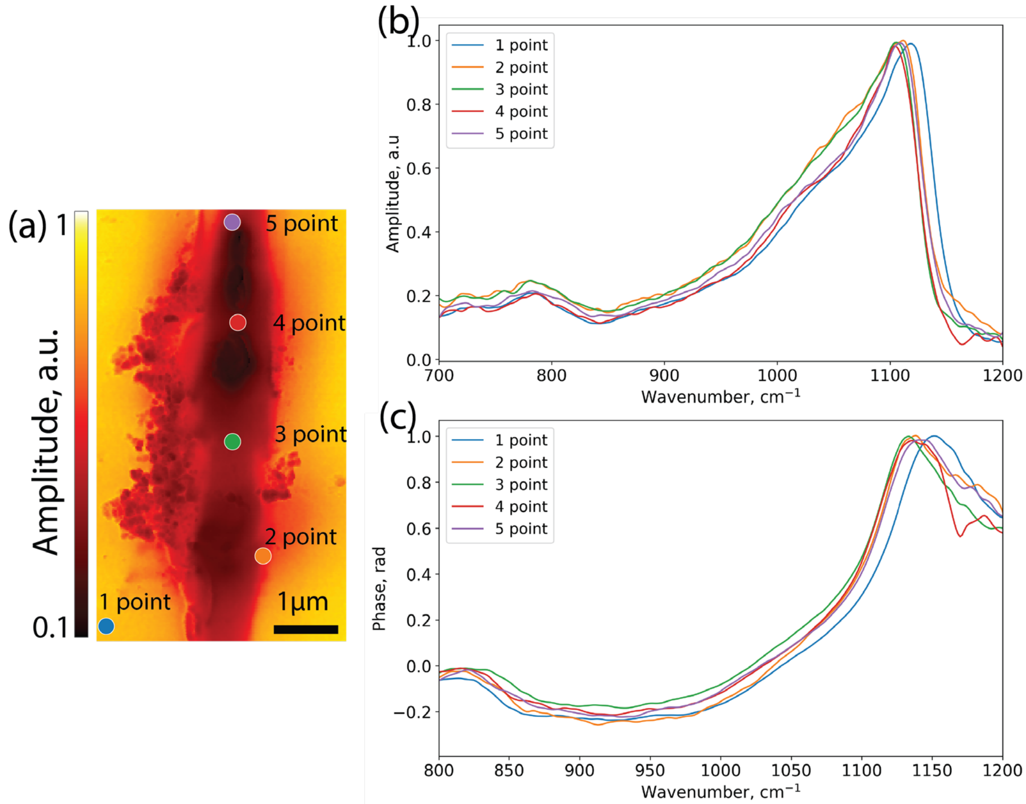

Fig. SI_1. The laser track written with 2 pulses per micron: (a) near-field amplitude map of the Si-O-Si asymmetric stretching mode at 1130 cm^{-1}; (b) synchrotron nano-FTIR amplitude and (c) phase spectra for measured points of pristine material (point 1) and irradiated area inside of the laser track (points 2-5).

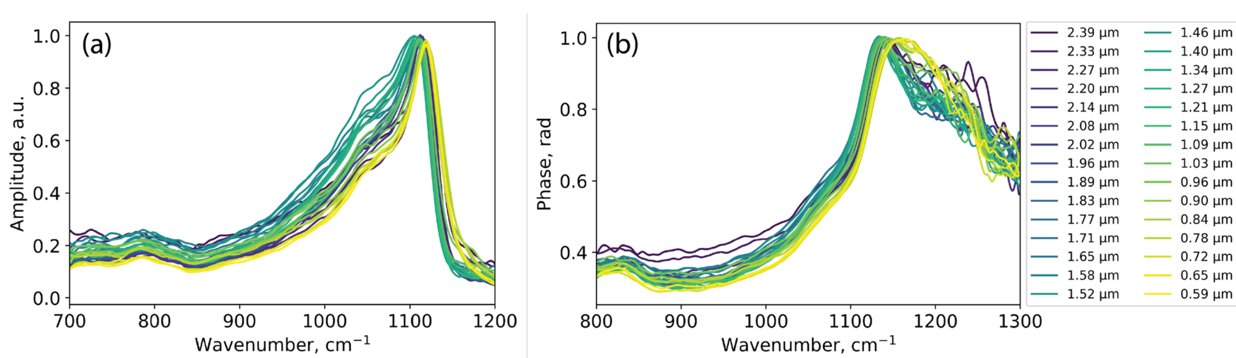

Fig. SI_2. Synchrotron nano-FTIR (a) amplitude and (b) phase spectra obtained along the line profile of the laser track written with 2 pulses per micron. This corresponds to the early birth of nanogratings. The legend indicates the X position of the spectrum on the line profile, as referenced in Fig. 3a-d of the manuscript. To avoid overloading the graph, only every second spectrum is plotted.

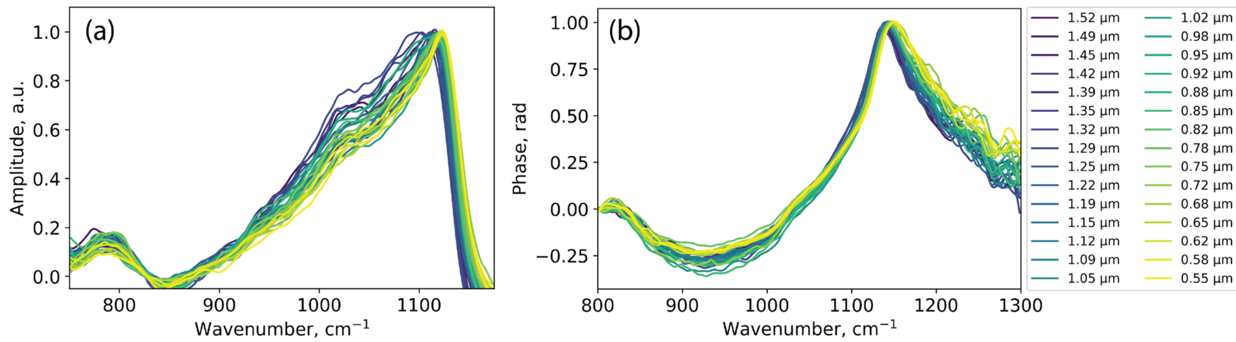

Fig. SI_3. Synchrotron nano-FTIR (a) amplitude and (b) phase spectra obtained along the line profile of the laser track written with 200 pulses per micron. This corresponds to well established nanogratings with a regular arrangement. The legend indicates the X position of the spectrum on the line profile, as referenced in Fig. 4 of the manuscript. To avoid overloading the graph, only every second spectrum is plotted.